PERGAMON

Energy Conversion and Management 42 (2001) 1919–1935

ENERGY CONVERSION & MANAGEMENT

www.elsevier.com/locate/enconman

Low NO_x combustion technologies for high temperature applications

Michael Flamme *

Gaswärme-Institut e. V., Hafenstraße 101, 45356 Essen, Germany

Abstract

Because of the high process temperature and the high temperature to which the combustion air is preheated, NO_x emissions from glass melting furnaces are extremely high. Even at these high temperatures, NO_x emissions could be reduced drastically by using advanced combustion techniques such as staged combustion or flameless oxidation, as experimental work has shown. In the case of oxy-fuel combustion, the NO_x emissions are also very high if conventional burners are used. Staged combustion achieves similar NO_x reductions.

Keywords: High temperature processes; Combustion air preheating; Oxy-fuel technology; NO_x reduction techniques

1. Introduction

Improvement of efficiency of combustion equipment is one of the key issues in order to reduce fuel consumption and CO_2 emission. Advanced systems for waste heat recovery, enforcement of heat insulation and combustion improvement have been developed. However, the increase in temperature of the combustion air raises the flame temperature and NO_x emissions rise sharply if conventional burners are used. At glass melting furnaces with process temperatures of up to 1600°C and more and air preheating temperatures of up to 1350°C the thermal NO formation increases drastically. Depending on the combustion technique used, NO_x emissions may therefore be extremely high. Potassium or sodium nitrate for refining particular grades of glass add to NO_x formation.

However, NO_x emissions have been limited in many countries because of their adverse impact on the environment. In Germany, maximum NO_x output levels for glass furnaces were first laid

* Fax: +49-201-3618238.

E-mail address: mflamme@gwi-essen.de (M. Flamme).

PII: S0196-8904(01)00051-6

down in the 1986 Clean Air Code which also contains a clause according to which further reductions are required in accordance with progress made in the state of the art.

2. Maximum NO_x emission levels in Germany

The Clean Air Code published in Germany in 1986 provides for maximum emissions for all pollutants. For many applications, though, the Code specifies minimum requirements as well as further reductions in accordance with the state of the art, because a substantial number of research and development projects were underway at the time when the Clean Air Code was published.

According to the Code, NO_x output from gas-fired side-port furnaces featuring regenerative combustion air preheating must not exceed 3500 mg/m^3. In the case of furnaces equipped with recuperators, NO_x emissions are limited to 1400 mg/m^3. The reference flue gas oxygen concentration is in both cases 8% (by volume).

However, retrofits are required according to the Clean Air Code to reflect technical progress in pollutant emission control. To make sure that this clause is interpreted in the same manner in all German states, an interstate pollution control committee agreed in 1991 on recommendations for the interpretation of this clause in the Clean Air Code. For glass furnaces, these recommendations quote a maximum NO_x emission level of 500 mg/m^3. They make reference to process modifications, low-NO_x burners and selective catalytic reduction to achieve these low levels [1].

As for the use of oxygen as an oxidant, the Clean Air Code does not publish any maximum NO_x levels. Although oxygen enrichment may increase the flue gas NO_x concentration sharply, theoretically, the process and any flue gas treatment plant must, according to the Code, be designed to comply with the requirements for combustion air. However, special arrangements may be allowed under licenses [2].

3. State of the art

Table 1 summarizes the NO_x emissions from glass furnaces as measured by Hüttentechnische Vereinigung der Deutschen Glasindustrie (HVG) [3]. The table lists for the different furnace designs NO_x concentrations typical of about 80% of the furnaces and the peak concentrations measured.

Substantially lower NO_x emissions can be achieved by novel combustion systems such as cascaded combustion [4] or through-port combustion [5]. Reburning (e.g. 3R method) [6] also reduces NO_x emissions. NO_x formation can further be curbed by the LoNO_x-Melter a new glass melting furnace with its completely new approach to glass melting. Cullet preheating and a reduction in the temperature to which the combustion air is preheated cut the NO_x concentration to below 500 mg/m^3 [7].

The alternative to curbing NO_x formation is, of course, post-formation clean up, for example, by selective catalytic or non-catalytic reduction [3]. However, the removal of NO_x from the flue gas is sometimes difficult because of the flue gas constituents. Further, it is, of course, more costly

Table 1
Overview of NO_x emissions from glass furnaces [3]

Type of glass furnace	Oil-fired (g/m^3)	Gas-fired (g/m^3)
Recuperative		
Typical range	0.4/1.4	0.4/1.6
Maximum		2.6
End-port furnace		
Typical range	1.0/2.4	1.4/3.0
Maximum range	4.6	
Side-port furnace		
Typical range	1.6/3.6	1.6/4.0
Maximum		5.6
Maximum incl. nitrate refining		7.0
Pot furnace		
Maximum incl. nitrate refining		4.0

both in terms of capital outlay and in terms of operating expenses to reduce NO_x emissions by removing the undesirable pollutants after they have formed.

Progress in the field of controlling NO_x formation has been remarkable in recent years. Recent research shows that NO_x output can be cut substantially even at a temperature of as much as 1600°C typical of glass industry applications.

4. Recent research

Gaswärme-Institut (GWI) has initiated different research programmes to explore methods of curbing NO_x emissions from high-temperature processes [8–10] using preheated combustion air or pure oxygen for oxidation.

4.1. NO_x control in processes using preheated combustion air

Basic research undertaken to reduce NO_x emissions in processes in which the combustion air is preheated to 1000°C and the furnace chamber temperature is 1200°C shows that new technologies such as staged combustion, flue gas recirculation and flameless oxidation (FLOX^{TM}) can curb the formation of NO_x substantially [9,11]. These technologies have in all cases been developed to reduce temperature peaks in oxygen-rich parts of the flame.

In the case of staged combustion, the combustion air is mixed with the fuel at different points. As heat is released more uniformly throughout the combustion chamber, NO_x formation is lowered notably. Fig. 1 shows a two-stage burner. As the secondary air is injected directly into the furnace chamber, flue gas is recirculated, increasing the NO_x reduction effect.

Flameless oxidation is another NO_x control technique. For flameless oxidation, the fuel and the air are both injected directly into the furnace chamber. They react downstream from the point of injection. That leads to very low flame temperatures and low oxygen partial pressures in the

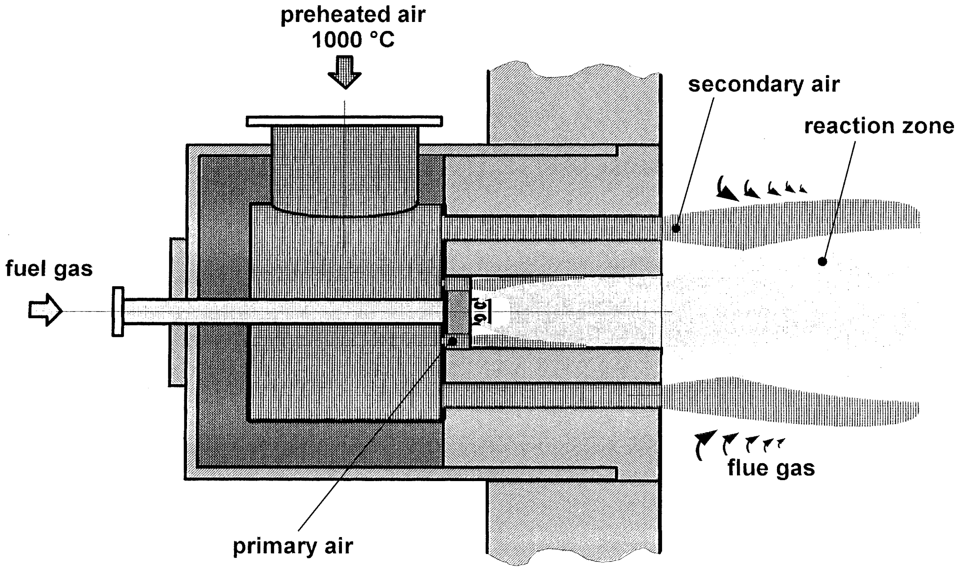

Fig. 1. Staged-combustion burner (burner 23).

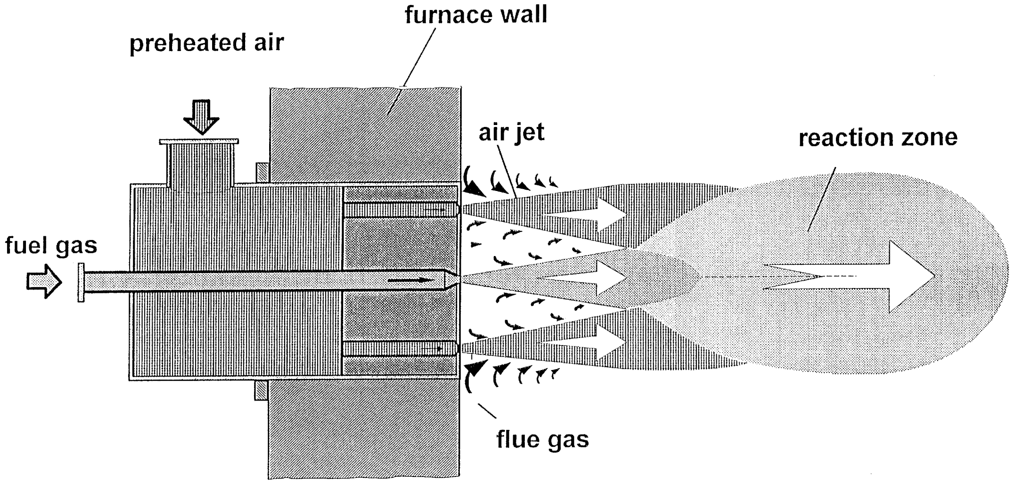

Fig. 2. Flameless oxidation burner (burner 25.1).

reaction zone and as a result of this to a low thermal NO formation. A flameless oxidation burner is shown in Fig. 2. Unlike staged combustion, flameless oxidation features no primary air. The energy required for ignition is provided by the recirculating flue gas. For this reason, the furnace chamber temperature must be at least 800–900°C for flameless oxidation.

Fig. 3 compares NO_x emissions from a conventional burner, a staged combustion burner and a FLOX^{TM} burner at different combustion air temperatures. For the tests, the furnace chamber temperature averaged 1200°C. As the graph demonstrates, staged combustion and flameless

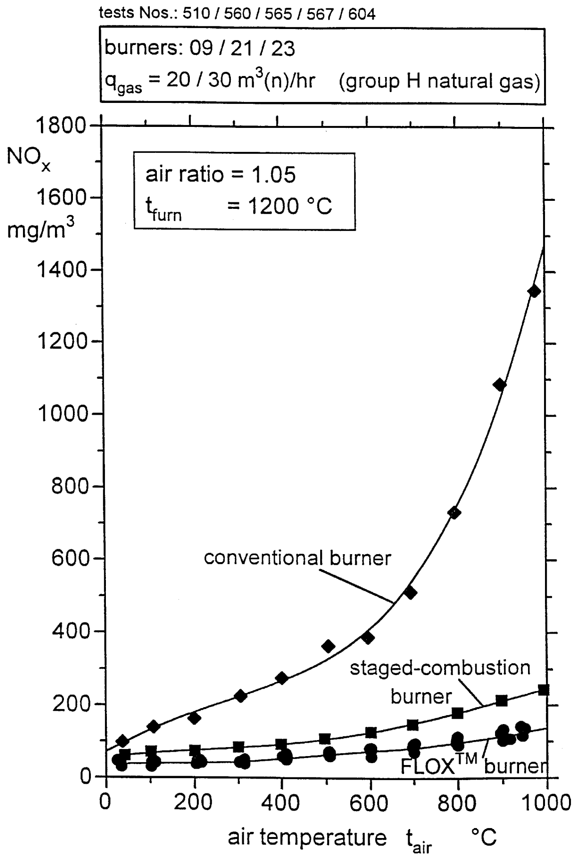

Fig. 3. NO_x concentration expressed as NO_2 (dry, 8% O_2) as a function of combustion air temperature t_{air}.

oxidation reduce NO_x output remarkably. For a combustion air temperature of 1000°C, for instance, the maximum NO_x concentration is 240 mg/m^3 for staged combustion and well below 180 mg/m^3 for flameless oxidation.

To obtain data on the peak reaction zone temperatures, temperatures and pollutant concentrations were measured (see Fig. 4) in the furnace chamber and, above all, in the vicinity of the burner [12]. In the case of flameless oxidation, the peak temperature in the reaction zone is merely about 100 K above the average furnace chamber temperature. Flue gas entering the reaction zone thus lowers temperature peaks considerably.

Optimization of the FLOX^{TM} system reduced NO_x emissions further. In the case of a regenerative flameless oxidation burner, it was even feasible to achieve an NO_x concentration of about 80 mg/m^3 in spite of a combustion air temperature of 1030°C and a furnace chamber temperature of 1200°C. As these data show, it is feasible to keep NO_x output very low by optimizing the combustion system.

The tests with process temperatures of up to 1200°C have been carried out at GWI test furnace 1. Fig. 5 shows the scheme of the furnace which is provided with an indirect cooling and heating system (radiant tubes) so that the furnace chamber temperature can be kept constant during the tests.

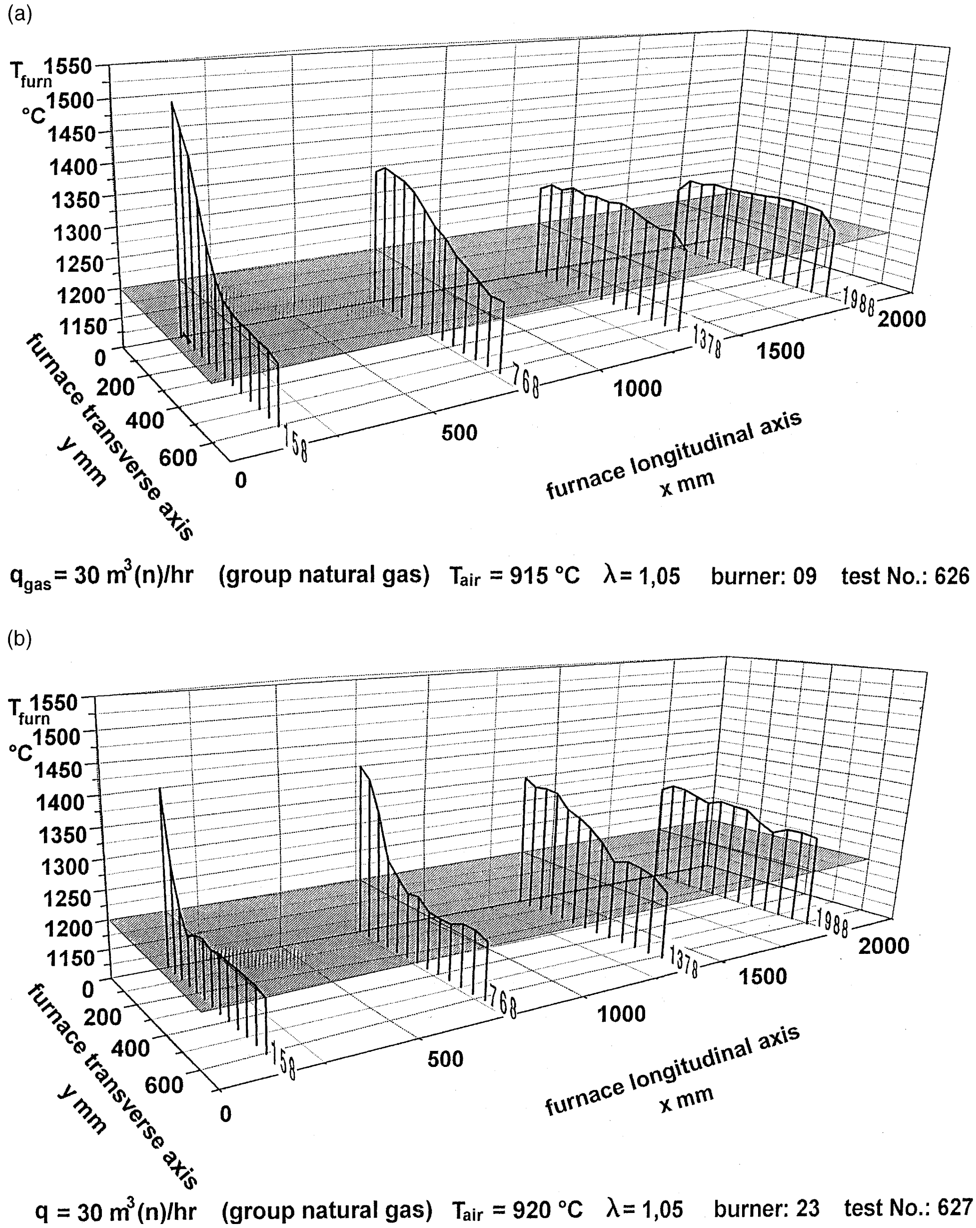

Fig. 4. Temperature distribution at GWI combustion chamber for different burners with air preheating of 920°C: (a) standard burner, (b) burner with air staging, (c) burner with flameless oxidation.

Apart from the actual combustion data, such as the combustion air temperature, the fuel/air ratio and the burner turn-down ratio, the furnace chamber temperature also has an influence on the total NO_x emissions. As the furnace chamber temperature rises, the temperature in the reaction zone increases. Fig. 6 plots NO_x concentration as a function of average furnace chamber temperature for a flameless oxidation burner [13]. For the tests, the laboratory furnace depicted in

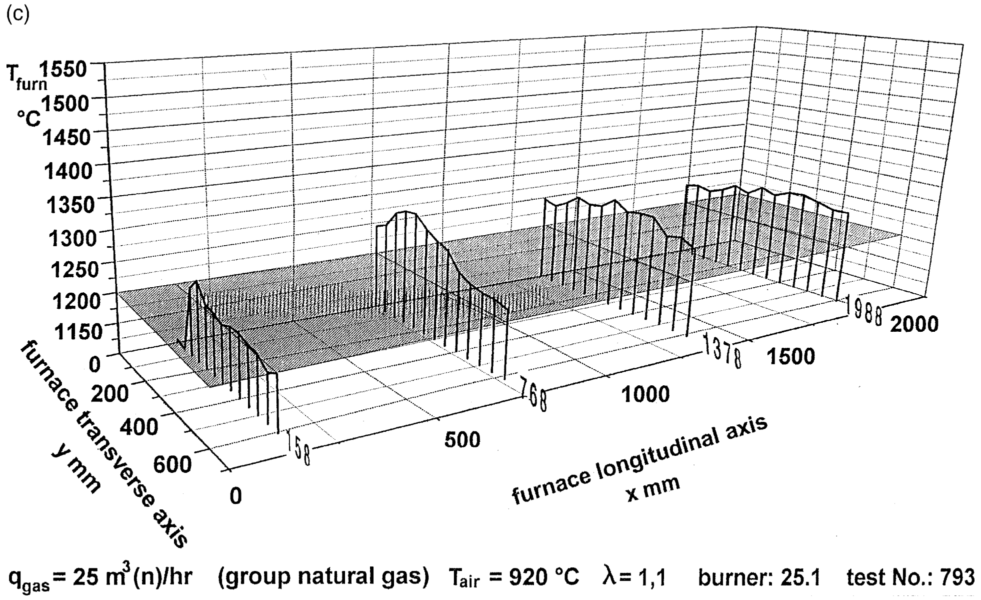

Fig. 4 (*continued*)

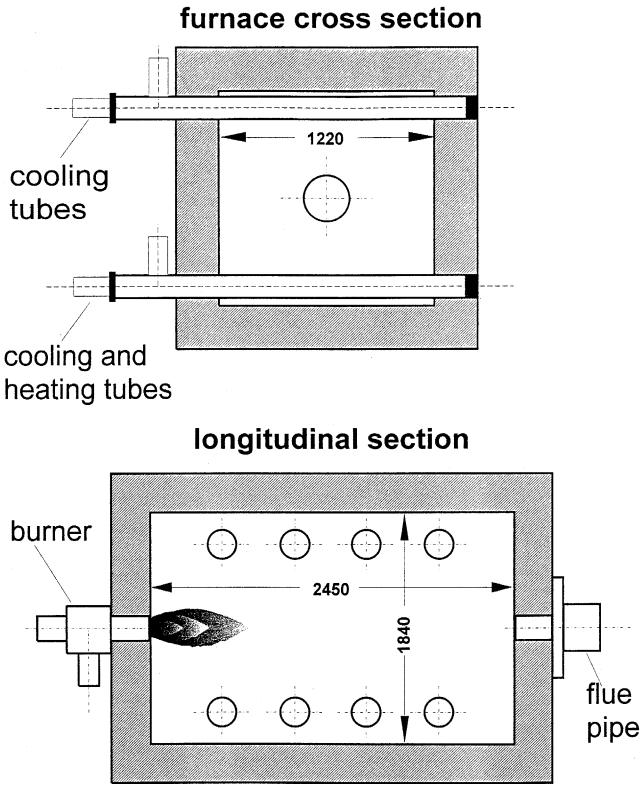

Fig. 5. Schematic of GWI laboratory furnace 1 (maximum 300 kW) for furnace chamber temperatures up to 1200°C.

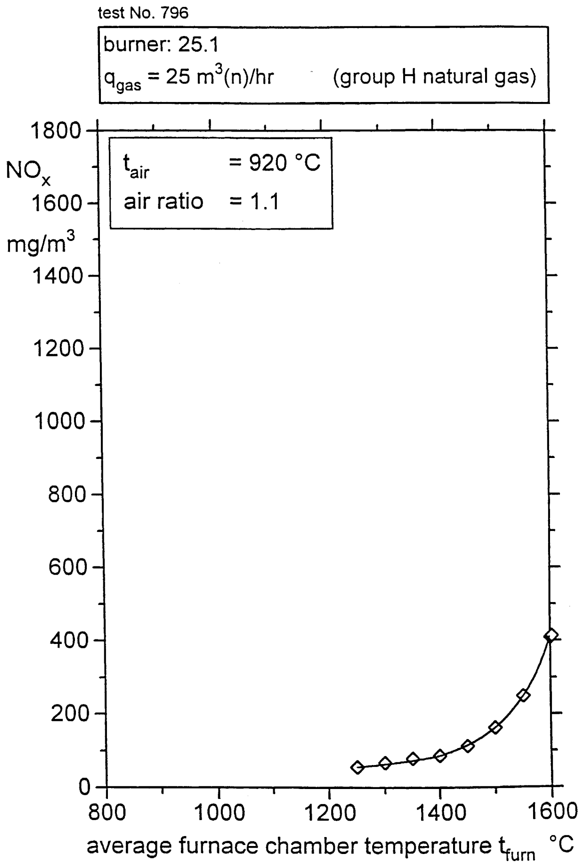

Fig. 6. NO_x concentration expressed as NO_2 (dry, 8% O_2) as a function of average furnace chamber temperature t_{furn} for a flameless oxidation burner (burner 25.1).

Fig. 7 was used. The combustion air temperature was 920°C. Under these conditions, the NO_x concentration rose from about 65 mg/m^3 at a furnace chamber temperature of 1200°C to about 410 mg/m^3 at 1600°C. NO_x concentrations quoted for the tests are referred to dry flue gas with an oxygen concentration of 8% (by volume). As these data demonstrate, the NO_x reduction potential is substantial even for high-temperature processes.

4.2. NO_x emissions of different burners for recuperative glass melting furnaces (unit melter)

To explore methods of controlling NO_x output from glass furnaces of the unit melter type featuring recuperative combustion air preheating without retrofitting flue gas treatment plants, GWI and Thyssengas GmbH decided to carry out a number of tests [14].

Various gas burners designed for heat inputs of 520 kW were tested. An electric air heater was installed to preheat the combustion air. In the tests, the combustion air temperature was 690°C. The resulting furnace surface loading was 167 kW/m^2 which is typical of glass tanks. The furnace chamber loading was 140 kW/m^3.

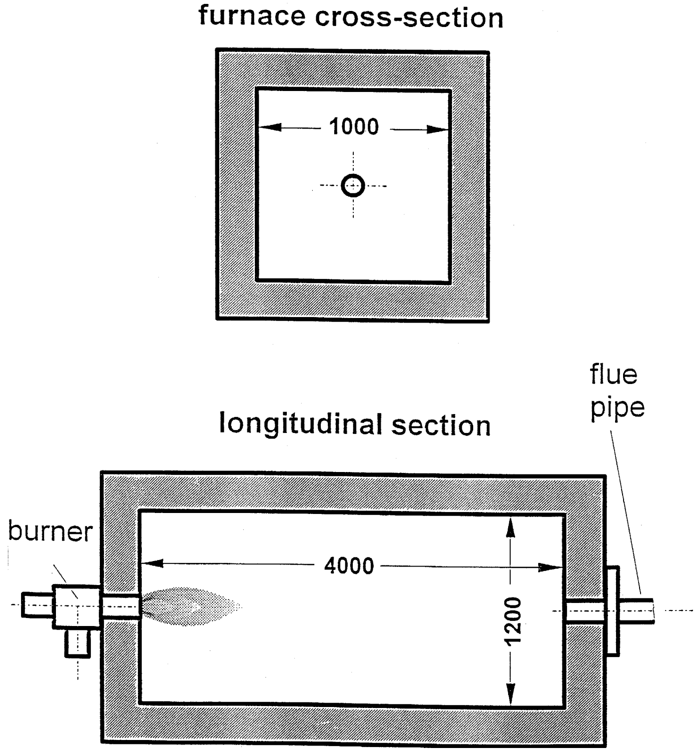

Fig. 7. Schematic of GWI laboratory furnace 2 (maximum 1.5 MW) for furnace chamber temperatures up to 1600°C.

4.2.1. Standard burner

To examine the influence of the mean furnace wall temperature on NO_x emissions, a burner (burner A) normally used in recuperative glass furnaces was tested. Fig. 8 is a schematic of the burner. The air ratio for the tests was 1.05.

NO_x measurements confirmed an over proportionate rise in NO_x concentration as the furnace wall temperature increased. The NO_x level peaked at 2.250 mg/m^3 [1] at a temperature of 1600°C (see Fig. 9). Glass furnace field measurements for burners of this type showed NO_x concentrations between 1200 and 1500 mg/m^3. One reason why the test furnace NO_x concentrations were higher is the compact design which prevents adequate flue gas recirculation in the furnace chamber. Secondly, the high NO_x level was due to the absence of the molten glass which would be at a lower temperature than the furnace floor. As the flame in the test furnace is completely surrounded by hot furnace walls, the flame temperature is higher than in a normal glass furnace.

To collect information on the flame length and temperature distribution in the furnace chamber, suction pyrometer measurements were carried out in the burner level. The distribution of the carbon monoxide concentrations in the furnace chamber is shown in Fig. 10. If burnout is

[1] All concentrations given as NO_2 (8% O_2, dry).

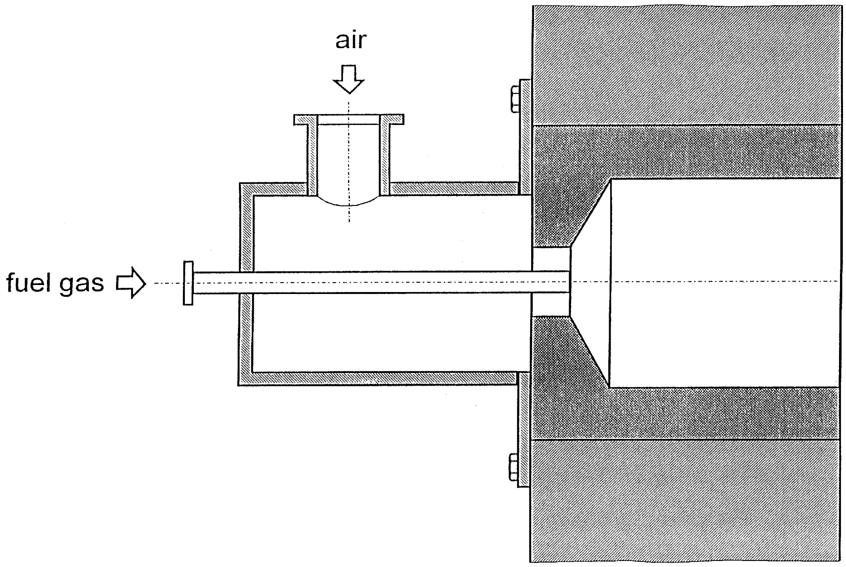

Fig. 8. Burner A.

equated with a CO concentration of 2000 ppmv, the length of the flame from burner A is approximately 1.3 m.

The burner A test data served as a reference line for evaluating the performance of the modified burners.

4.2.2. Staged-combustion burner

Basic research shows that combustion air staging can reduce NO_x emissions further. It is crucial though , for heat to be removed from between the primary and the secondary combustion areas. This dissipation can be achieved by moving the burnout zone into the furnace chamber. Aspiration of flue gas from the furnace chamber into the secondary air lowers NO_x emissions by another percentage [15]. For staged combustion in accordance with the principles described, the burner brick was modified as shown in Fig. 11 (burner C). This burner brick geometry splits combustion air flow into equal primary and secondary air flows.

The effect of the modification on NO_x emission is depicted in Fig. 9. While mean furnace wall temperature was 1600°C, NO_x output was only about 750 mg/m^3 equivalent to a reduction of 1500 mg/m^3 (67%) by comparison with burner A (the standard burner). Considering that the rate of NO_x formation in the test furnace is higher than in a real glass furnace, it appears feasible to achieve the State Pollution Control Committee objective of 500 mg/m^3 using this burner. This optimistic prediction requires confirmation by testing the new burner system in a glass tank, though.

As Fig. 12 shows, the flame length of burner C is about 2.6 m, while the diameter of the combustion area is approximately 0.6 m. The comparison of the flame length produced by burners

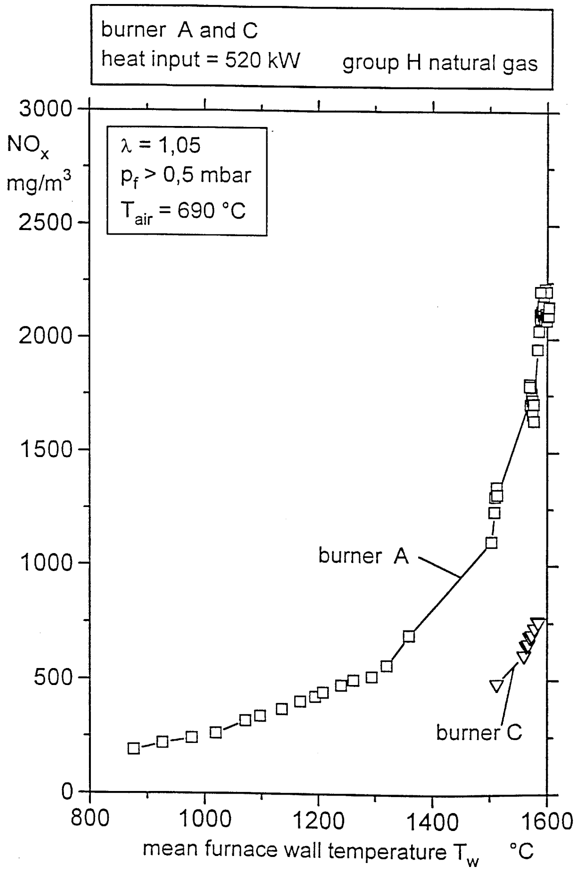

Fig. 9. NO_x emission from burner A and C as a function of mean furnace wall temperature T_w.

A and C demonstrates that the combustion zone of a staged-combustion system can be relatively narrow if the secondary air nozzle arrangement is optimized.

4.3. NO_x control in processes using pure oxygen

If pure oxygen is used for combustion, the oxidant carries practically no nitrogen which could form NO_x. The nitrogen that is converted is a constituent of the fuel. The nitrogen content of natural gas varies substantially, though, and may range between a few percent and as much as 14% (Dutch natural gas). Such a high nitrogen content would cause substantial NO_x emission, if the fuel were burnt in a conventional natural gas/oxygen burner. Further, nitrogen may enter the combustion zone if a furnace is not tight, increasing the NO_x formation rate. In tests carried out to investigate the correlation between the fuel gas nitrogen concentration and NO_x emissions, the pressure in the furnace chamber was therefore kept above atmospheric pressure.

To analyze the effect of the nitrogen content of the flue gas on NO_x output, different CH_4/N_2 mixtures were burnt [16]. As Fig. 13 shows, the flue gas NO_x concentration rises sharply, as the

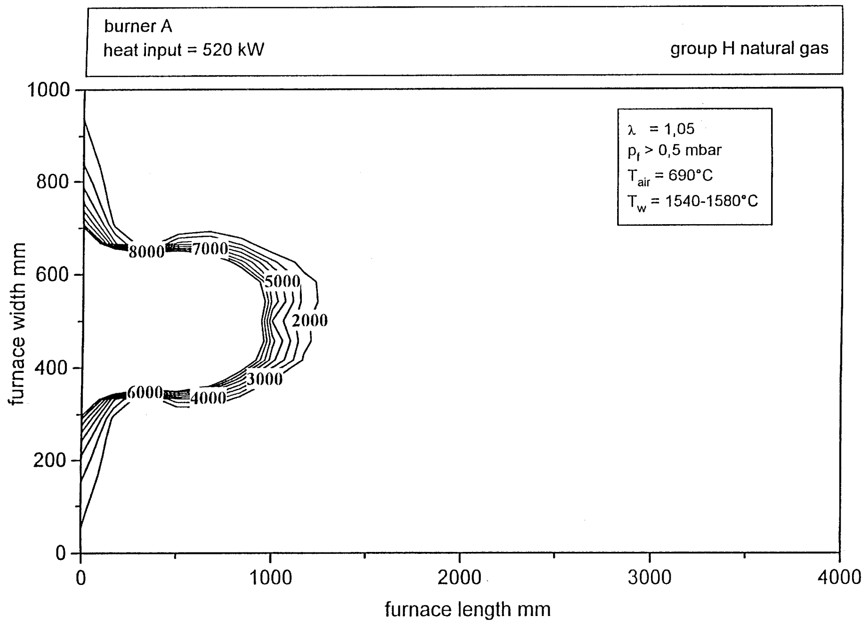

Fig. 10. Burner A CO concentration in the burner level in the furnace chamber.

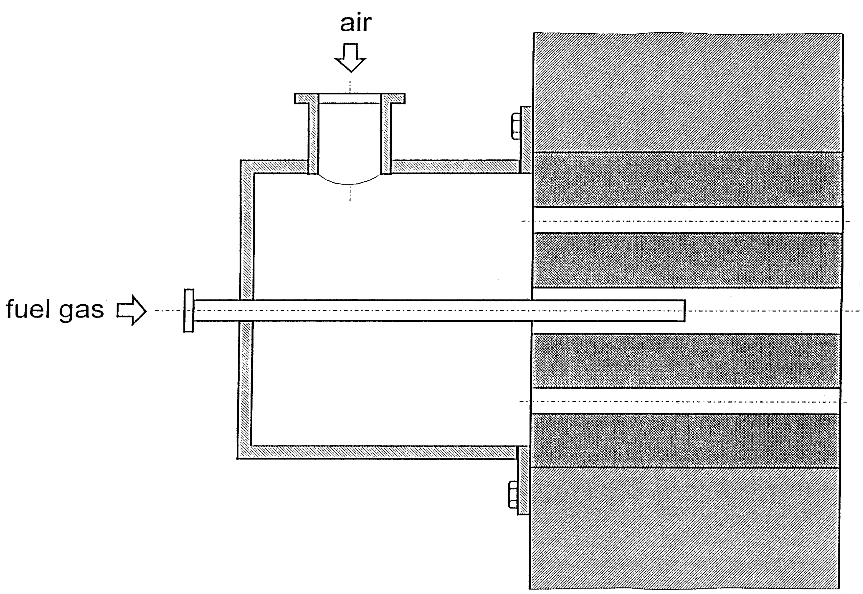

Fig. 11. Burner C.

nitrogen concentration of the fuel gas increases. In the test the fuel was combusted by a parallel-jet nozzle mixing burner at two different oxidant ratios.

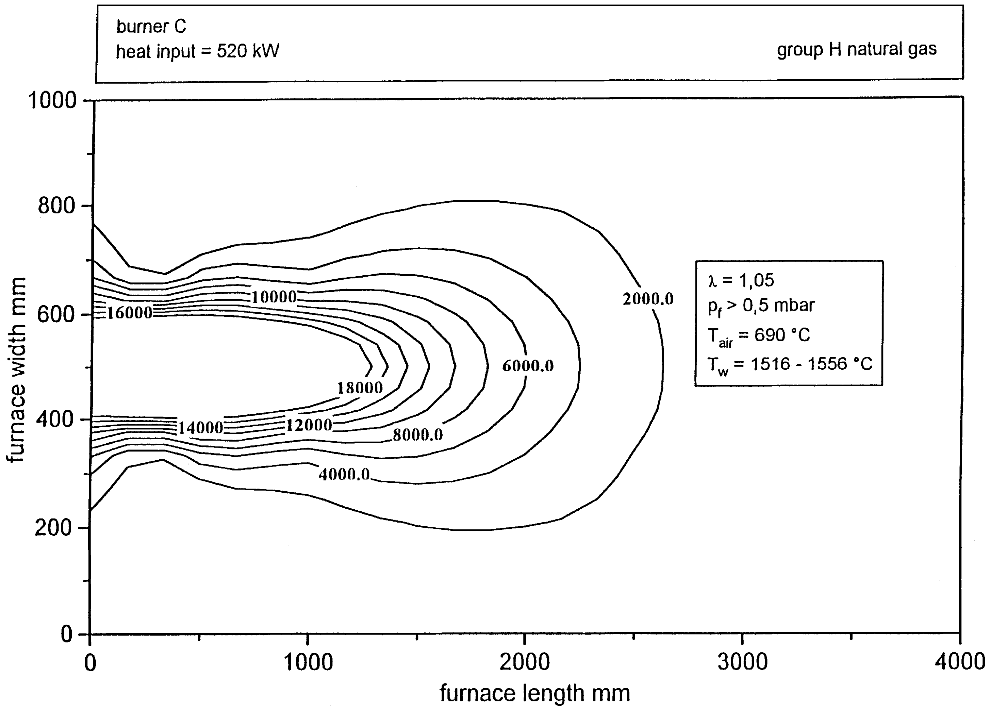

Fig. 12. Burner C CO concentration in the burner level in the furnace chamber.

In further tests, NO_x reduction by staged combustion and flue gas recirculation was examined [10]. For the staged combustion work, the pilot burner depicted in Fig. 14 was built. Axial displacement of the fuel gas lance varies the primary oxygen share. The lance position I is thus a measure of the primary oxygen share in the total oxygen. If I = 0, no primary oxygen is injected. As the lance is withdrawn from the burner mouth, the primary oxygen share increases.

Fig. 15 correlates the lance position I and the NO_x concentration measured. The NO_x content of the flue gas decreases from about 960 mg/m^3 for I = 10 mm to 100 mg/m^3 for I = 0. In the tests, the average furnace chamber temperature was 1200°C. The furnace chamber was lined by a ceramic fiber material (see Fig. 5).

To examine the effect of the furnace chamber temperature on NO_x output from this burner, a further series of tests without primary oxygen injection was carried out, using the laboratory furnace shown in Fig. 7. As Fig. 16 shows, NO_x output rose slowly as the furnace chamber temperature was increased, going up from about 40 mg/m^3 to approximately 340 mg/m^3. The relatively low NO_x concentration at an average furnace chamber temperature of 1200°C (when compared with Fig. 15) is attributable to the low furnace wall temperature. The furnace chamber depicted in Fig. 7 is lined by refractory concrete. For this reason, the wall temperature rises more slowly than in the case of a furnace lined by ceramic fiber material during the heating up period. If the furnace chamber temperatures reaches 1600°C there is no big difference between furnace wall temperature and furnace chamber (gas) temperatures. The work showed that staged combustion can substantially lower NO_x emission when oxygen is used as an oxidant.

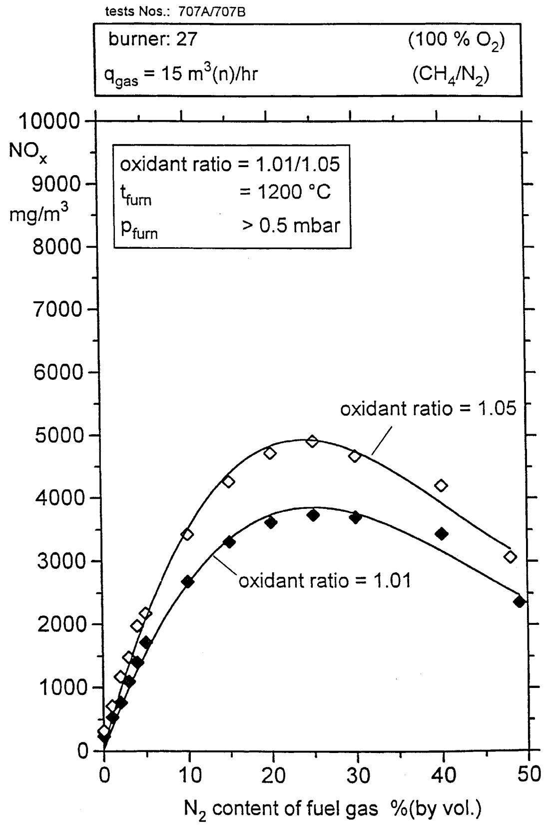

Fig. 13. NO_x concentration (dry, 8% O_2) as a function of N_2 content of fuel gas.

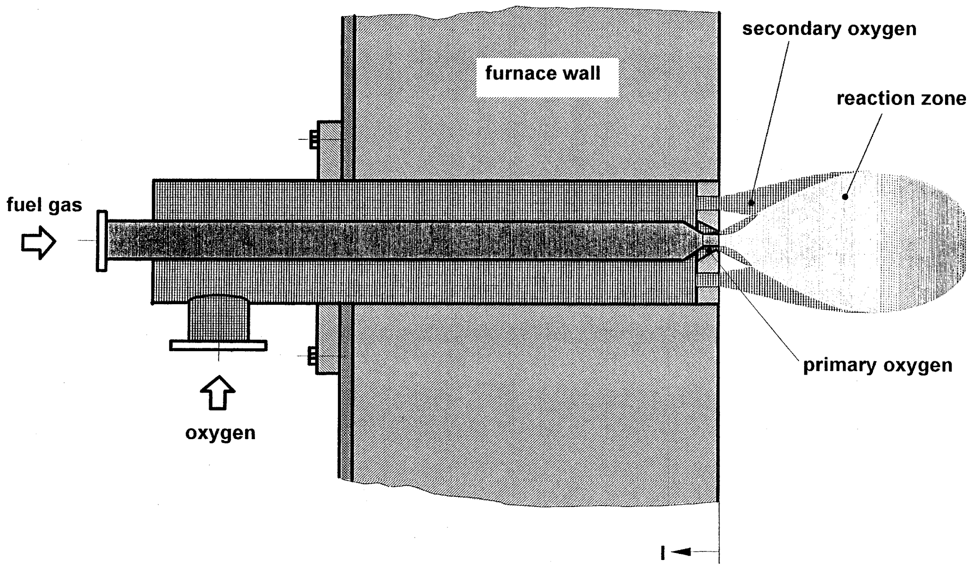

Fig. 14. GWI pilot burner to test staged oxygen combustion (burner 34.4).

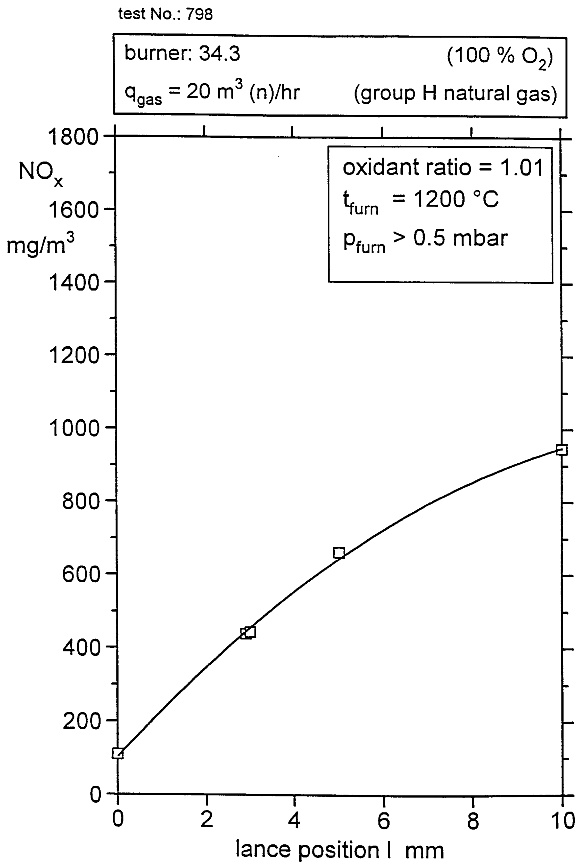

Fig. 15. NO_x concentration expressed as NO_2 (dry, 8% O_2) as a function of lance position I.

Apart from the fact that these new combustion techniques could solve the NO_x problem for glass melting furnaces with recuperative air preheating these advanced combustion systems offer new approaches for a lot of other combustion equipment. With the new burner generation it is possible to increase the air preheating temperature without an increase in NO_x formation. The uniformity in temperature distribution allows to raise the mean furnace chamber temperature without a risk of overheating in the near burner region. This leads to an improvement of heat transfer and high energy savings [17].

5. Prospects

Work by GWI showed that NO_x emissions can be reduced drastically at least under laboratory conditions. Further studies investigating heat transfer at a temperature of 1600°C will have to show whether these advances low NO_x combustion techniques could be used for glass melting furnaces with recuperative air preheating. These studies will be followed up by actual field test.

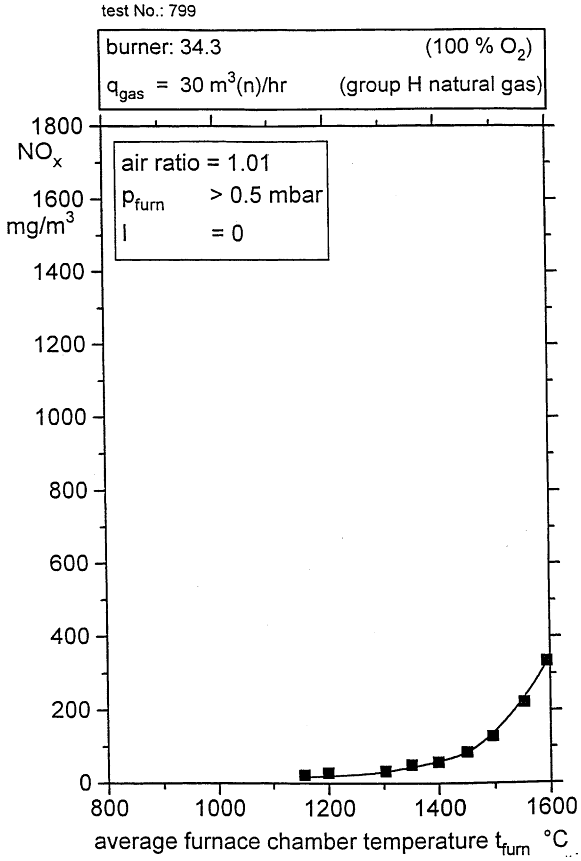

Fig. 16. NO_x concentration expressed as NO_2 (dry, 8% O2) as a function of average furnace chamber.

Acknowledgements

The work presented in this paper was sponsored by the German Ministry of Research and Technology, the German gas industry, the German Confederation of industrial research associations, N.V. Nederlandse Gasunie and Ruhrgas.

References

[1] Lange M. Neue Vorschriften und Maßnahmen zur Luftreinhaltung, Energieeinsparung und Reststoffverwertung bei Industriefeuerungen. Gaswärme Internat 1992;41:10.
[2] Davids P, Lange M. TA Luft '86. Technischer Kommentar. Düsseldorf: VDI Verlag; 1986.
[3] Kircher U. NO_x-Emissionen und Stand der Minderungstechnik. Proceedings of the HVG Course on the Reduction of Solid Particle and Gaseous Pollutant Emissions from Glass Melting Furnaces. Nürnberg, 15, 16 November, Duisburg, 18, 19 November, 1993.
[4] Pieper H. Die großen U-Flammenwannen mit 100 m^2 und mehr Schmelzfläche. Proceedings of Glastechnische Tagung, Königswinter, 1993.

[5] Schwarz G. Feuerungstechnik für Glasschmelzwannen gestern und heute. Proceedings of Glastechnische Tagung, Bad Salzdetfurth, 1994.

[6] Friedrich H. 3R-Verfahren (Poster). Proceedings of Glastechnische Tagung, Bad Salzdetfurth, 1994.

[7] Pieper H, Patzer T, Becher J. Ökologischer und ökonomischer Vergleich zwischen einer modernen U-Flammenwanne und der zweiten Generation des Sorg LoNOx-Melters. Proceedings of Glastechnische Tagung, Bad Salzdetfurth, 1994.

[8] Flamme M. Minimierung der Stickstoffoxidbildung bei der Verbrennung von Erdgas mit starker Luftvorwärmung in Hochtemperaturprozessen. Thesis, Ruhr-Universtität Bochum, 1989.

[9] Flamme M. Energieeinsparung und Schadstoffminderung bei Industrieöfen der Stahl- und NE-Metallindustrie durch Weiterentwicklung und Optimierung von Regeneratorbrennern. GWI report no. 9753, 1993.

[10] Flamme M, Brune M, Haep J, Kremer H. NO_x-Emission bei der Erdgasverbrennung mit sauerstoffangereicherter Verbrennungsluft. GWI report no. 9912, 1994.

[11] Wünning JA, Wünning JG. Brenner für die flammenlose Oxidation mit geringer NO-Bildung auch bei höchster Luftvorwärmung. Gaswärme-Internat 1992;41:10.

[12] Flamme M, Haep J. Einsatz neuer Brenntechniken. GWI report no. 9434, 1993.

[13] Haep J, Flamme M. Untersuchung der NO_x-Emission bei flammenloser Oxidation und Ofenraumtemperaturen bis 1600°C und Ermittlung des Wirkungsgrades eines FLOX-Brenners im Vergleich zu einem konventionellen Brenner. GWI report no. 9963, 1994.

[14] Flamme M, Ditscheid U. NO_x-arme Brennertechnik für Glasschmelzwannen mit rekuperativer Luftvorwärmung. Gaswärme Internat 1996;45:3.

[15] Brenner für Hochtemperaturprozesse und Verfahren zu seinem Betreiben. Patent application no. DE 4130922A1, 17 September, 1991.

[16] Flamme M, Haep J. Influence of different nitrogen amounts in fuel gas on NO_x emission in case of pure oxygen firing. GWI report no. 9391, 1993.

[17] Tanaka R. New progress of energy saving technology towards the 21st century. Frontier of combustion and heat transfer technology. Proceedings of the 11th Members Conference of the International Flame Research Foundation. The Netherlands: Noordwijkerhout; 1995.